

Adatom incorporation and step crossing at the edges of 2D nanoislands

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Adatom incorporation into the “faceted” steps bordering the 2D nanoislands is analyzed. The step permeability and incorporation coefficients are derived for some typical growth situations. It is shown that the step consisting of equivalent straight segments can be permeable even in the case of fast edge migration if there exist factors delaying creation of new kinks. The step consisting of alternating rough and straight segments may be permeable if there is no adatom transport between neighboring segments through the corner diffusion.

Surface evolution in epitaxial growth is known to be highly sensitive to details of interaction of adatoms with the monoatomic steps.[1] In the continuous approach such details should be taken into account in the incorporation and step permeability coefficients appearing in the boundary conditions for the surface diffusion equation. However, it is still a common praxis to treat the kinetic coefficients as the phenomenological Arrhenius-like constants. One can show that in many cases, especially when the adatom incorporation into the step is limited by the process of creation of kinks at the step edge [2, 3], such a simplified approach is not correct.

Recently we have proposed a method to *derive* the incorporation and step permeability coefficients of vicinal steps aligned along high symmetry directions.[4] The aim of the present paper is to extend this approach to construct the kinetics coefficients of the edges of 2D nanoislands. The specific shape and small size of the 2D islands give rise to the peculiarities in the kinetics of adatom incorporation and crossing the island edge as compared to the case of the “infinite” vicinal steps. So, the length of the edge of a 2D island may be less than or comparable with the average distance between neighboring kinks at the edge of a vicinal step situated at the same surface under the same growth conditions. Then it is highly probable for the 2D island to be of a strongly polygonized shape, i.e. to be bordered by a “faceted” step containing few or even no kinks. This is exactly what was observed e.g. in the scanning tunnelling microscopy studies of growth of Si and Ge on the Si(111) and (001) surfaces.[5]

The description of adatom incorporation into the “faceted” islands inevitably involves analysis of two processes - creation of kinks at the step “facets” and the “interfacet” material transport. Both processes have been addressed in the literature only in the limiting cases of irreversible attachment of the terrace adatoms to the edge or weak migration of the edge adatoms.[2, 6, 7] In the present paper we get rid of those simplifying assumptions and derive the incorporation and step permeability coefficients for two typical growth situations - the 2D islands with equivalent step segments and the 2D islands with alternating atomically straight and rough step segments (as, e.g. the Si islands on the Si(111)-7x7 and Si(001)-2x1

surfaces, respectively).

We will consider propagation of a step segment which length L is less than the average distance between kinks L_k at the infinite step considered at the same growth conditions. An adatom attached to such a segment has four possibilities (Fig. 1): (1) to detach from the segment back to the terrace from which it came or to the adjacent terrace (the latter means the crossing of the step); (2) to leave the segment by rounding the island corners; (3) to meet another adatom or an unstable cluster at the same segment and in that way to take part in the 1D nucleation process. After appearance of the 1D island the adatom can (4) incorporate into one of two kinks at its ends.

As the result, the row-by-row growth process sketched in Fig. 1 will take place. It includes appearance of the 1D island at the straight (without kinks) step segment after the expectation time t_{nuc} and its spreading along the step edge during the mean time t_{gr} . It is essential that no other 1D islands appear during the time t_{gr} . As the crystalline row along the step segment completes, the 1D nucleation and formation of the new crystalline row start again.

If the adatoms do not migrate along the step edge then $t_{nuc} = 1/LJ$, where J is the 1D nucleation rate per atomic site at the step.[2, 6] In this case $t_{nuc} \gg t_{gr}$ and the maximum of the step permeability is achieved, the relevant expressions for the kinetic coefficients can be found in Ref. 8. In the present paper we are interesting in the opposite limit of fast edge migration, when the traversal time required for an adatom to visit all sites at the edge $t_{tr} \sim L^2/D_e$ is much less than the mean time

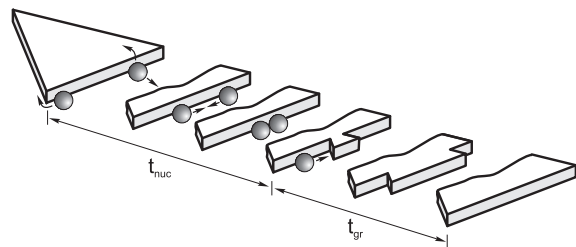


FIG. 1: Schematic of atomic processes at the edge of the 2D island during the row-by-row growth process.

interval between subsequent attachments of the terrace adatoms to the edge $\Delta t = 1/L(k_{le}^+ n_l + k_{ue}^+ n_u)$ and the mean time $t_{res} = 1/(k_{el}^- + k_{eu}^-)$ that an adatom spends at the straight step edge before detachment, where D_e is the edge diffusion coefficient, $k_{l(u)e}^+$ and $k_{el(u)}^-$ are the attachment (+) and detachment (-) rate constants, and n_l and n_u are the concentrations of adatoms on the lower (l) and upper (u) terraces in the close vicinity to the 2D island edge. Bearing in mind comparatively low growth temperatures we neglect detachment of atoms embedded into the straight step as well as into the kink and corner sites.

Using the above picture of elementary processes we calculate the net flux (the exchange rate) of adatoms between the island edge and the adatom gas at the lower terrace averaged over the edge length and the period $t_{1D} = t_{nuc} + t_{gr}$ of formation of the crystalline row along the edge

$$g_l = \frac{1}{L} \int_0^L \frac{1}{t_{1D}} \int_0^{t_{1D}} j_l(x, t) dx dt. \quad (1)$$

Here x is the coordinate along the island edge and $j_l(x, t)$ is the local net flux. In the limit of fast edge migration an adatom which attaches to the edge segment during the time interval t_{gr} has no chances to detach from this segment (it may however to leave the segment via corner rounding). Then one can express $j_l(x, t)$ as

$$j_l(x, t) = \begin{cases} k_{le}^+ n_l - k_{el}^- n_e(x), & \text{within the time interval } t_{nuc} \\ k_{le}^+ n_l, & \text{within the time interval } t_{gr} \end{cases}$$

where we assume that the concentration of adatoms on the terrace does not change considerably during the time interval t_{1D} (the concentrations of the terrace adatoms n_l and n_u are considered as unknown variables for which the usual quasi steady-state approximation holds).

The integration gives

$$g_l = (1 - \tau_k)(k_{le}^+ n_l - k_{el}^- \bar{n}_e) + \tau_k k_{le}^+ n_l \quad (2)$$

where $\tau_k = t_{gr}/(t_{gr} + t_{nuc})$ is the fraction of time when every adatom attaching to the edge contributes to the formation of the crystalline layer along one of the edge segments and

$$\bar{n}_e = \frac{1}{L} \int_0^L n_e(x) dx$$

is the mean concentration of the edge adatoms. The concentration $n_e(x)$ is found as the solution of the edge diffusion equation

$$D_e \frac{d^2 n_e}{dx^2} - (k_{el}^- + k_{eu}^-) n_e(x) + k_{le}^+ n_l + k_{ue}^+ n_u = 0 \quad (3)$$

with the boundary conditions describing incorporation of the edge adatoms at the kink sites or/and leaving the

edge segment at the island corners. In the following we summarize our major results for some typical growth situations.

The 2D islands with equivalent step segments.

Assuming fast migration of the edge adatoms around the island corners (this process has a high probability e.g., in the case of triangular 2D Si islands on the Si(111)-7x7 surface)[5] we adopt the periodical boundary conditions at the corners. In this case the concentration of the edge adatoms does not depend on x and is given by $n_e = (k_{le}^+ n_l + k_{ue}^+ n_u)/(k_{el}^- + k_{eu}^-)$. Substituting this expression into Eq. (2) we obtain the exchange rate g_l in the standard form[9]

$$g_l = \nu_l(n_l - \bar{n}) + \nu_p(n_l - n_u). \quad (4)$$

The first term in the right part of Eq. (4) is the flux of adatoms incorporating into the kinks at the island edge and the second term is the flux of adatoms crossing the edge without visiting the kinks, ν_l and ν_p are the incorporation and step permeability coefficients, respectively. The coefficients are given by

$$\nu_l = \tau_k k_{le}^+; \quad \nu_p = \frac{(1 - \tau_k) k_{le}^+ k_{eu}^-}{k_{eu}^- + k_{el}^-}. \quad (5)$$

Similar expression for the flux g_u of adatoms leaving the upper terrace and relevant kinetic coefficients may be obtained by the substitution l for u and *vice versa* in Eqs. (4) and (5).

The ability of the adatoms from the lower terrace to cross the edge and thus climb up the 2D island top may be characterized by the ratio

$$\eta_l = \frac{\nu_p}{\nu_l} = \frac{t_{nuc} k_{eu}^-}{t_{gr} (k_{eu}^- + k_{el}^-)}. \quad (6)$$

As can be seen from Eq. (6), the edge segment may be permeable ($\eta_l \gg 1$) if its propagation is limited by the kink creation ($t_{gr} \ll t_{nuc}$) even if migration of the edge adatoms is fast.

The 2D islands with inequivalent step segments.

We have considered the case of the 2D island with alternating atomically straight and rough edge segments as e.g., SA and SB edge segments of the rectangular 2D Si islands on the Si(100)-2x1 surface. Here the probabilities for an adatom to cross the straight segment or incorporate into it are both affected by the ability of the edge adatom to travel around the step corners. Assuming that the adatom does not return back from the neighboring rough segments we get

$$g_l = \nu_l(n_l - \bar{n}) + \nu_p(n_l - n_u) + \nu_{ec} n_l, \quad (7)$$

where the term $\nu_{ec} n_l$ is the flux of adatoms attaching to the segment and leaving it via the corner rounding. The kinetic coefficients appearing in Eq. (7) are written in the form

$$\nu_l = \kappa \tau_k k_{le}^+;$$

$$\nu_p = \frac{(1 - \tau_k)[1 - f_c(q_L)]k_{le}^+k_{eu}^-}{k_{eu}^- + k_{el}^-};$$

$$\nu_{ec} = [(1 - \kappa)\tau_k + (1 - \tau_k)f_c(q_L)]k_{le}^+,$$

where

$$f_c(q_L) = \frac{\tanh(q_L)}{q_L [1 + 2q_L \tanh(q_L) D_e / (k_{ec} L)]}$$

is the probability that an adatom, attached to the step segment containing no kinks, will leave the segment via the corner rounding before detachment, q_L is the ratio of the segment length to the mean length of the adatom migration along the infinite step, k_{ec} is the rate constant for corner rounding and κ ($0.5 \leq \kappa \leq 1$) is the probability that the edge adatom will find the kink when the latter is present at the given step segment.

The permeability ratio in this case is given by

$$\eta = \frac{\nu_p}{\nu_l + \nu_{ec}} = \frac{(1 - \tau_k)[1 - f_c(q_L)]k_{eu}^-}{[\tau_k + (1 - \tau_k)f_c(q_L)](k_{el}^- + k_{eu}^-)}.$$

Here the neighboring step segments act as a pair of kinks settled at a short ($L < L_k$) distance. This diminishes crossing the step by the terrace adatoms. Our calculations give that the step segment may be permeable only if the energy barrier for the corner rounding is greater than the barrier E_e for the edge migration by $\Delta E_{ec} > E_{es}^- - E_e - k_B T \ln(L/2)$, where E_{es}^- is the smallest from the barriers for detachment to the upper and lower terraces.

In conclusion, we have derived the incorporation and step permeability coefficients for two typical growth situations involving the 2D islands bounded by the “faceted” steps. It has been shown that adatom incorporation into such islands has some peculiarities which are reflected by the structure of the kinetic coefficients.

It should be noted that in spite of the linear form of Eqs. (4) and (7) the exchange rates are in general the *non-linear* functions of the adatom concentrations in the vicinity of the island edge because propagation of the edge segments involves a non-linear process of formation of kinks by the non-equilibrium 1D nucleation mechanism. The coefficients ν_l and ν_p are, in fact, the functions of the adatom concentrations n_u and n_l , and they reduce to the Arrhenius-like constants only when the system is close to equilibrium or in the case of irreversible

attachment of the adatoms to the island edge (i.e. when $\tau_k = 1$).

To apply our model to the particular system of interest one needs to specify the characteristic time scales t_{gr} and t_{nuc} . Evidently, $t_{gr} \sim 1/(k_{le}^+n_l + k_{ue}^+n_u)$ with the coefficient of proportionality (order unity) depending on the 2D island geometry and intensity of the corner rounding processes. The nucleation time t_{nuc} is the inverse of the nucleation rate which can be calculated with the statistical theory of the island-on-island nucleation.[10] An application of the outlined strategy to modeling of the mass-transport during the formation of the multilayer Ge nanoislands on Si(111)[11] can be found elsewhere.[12]

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